Modeling Stratospheric Constituents: Reactive Species That Regulate Ozone

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Photochemical loss of stratospheric ozone occurs primarily by catalytic cycles whose rates are limited by the concentration of OH, HO2, NO2, Clo, and/or BrO as well as the concentration of either atomic oxygen or of ozone itself. Once the concentrations of these gases are established, the photochemical loss rate of O3 depends on the rate coefficient of only a handful of key reactions. We have developed a method for testing our understanding of stratospheric ozone photochemistry by comparing measured and modeled concentrations of reactive hydrogen, nitrogen, chlorine and bromine radicals using a photochemical steady state model constrained by observed concentrations of long-lived precursors (e.g., NOy, Cly, Bry, O3, H2O, CH4) and environmental parameters such as ozone column, reflectivity, and aerosol surface area. We will show based on analyses of observations obtained by aircraft, balloon, and satellite platforms during the POLARIS campaign that our overall understanding of the processes that regulate these radical species is very good. The most notable current discrepancies are the tendency to underestimate observed NO2 by 15 to 30% for air masses that experience near continuous solar illumination over a 24 hour period and the tendency to underestimate observed OH and HO2 by about 10 to 20% during midday and by much larger amounts at high solar zenith angle (SZA > 85). Possible resolutions to these discrepancies will be discussed. This study was carried out in close collaboration with many members of the POLARIS science team.